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(54) RESIN COMPOSITION

HARZZUSAMMENSETZUNG

COMPOSITION DE RÉSINE

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(56) References cited:
WO-A2-2020/100102
JP-A- 2007 277 469
JP-A- 2014 032 966
KR-A- 20100 065 731
KR-A- 20190 113 585
US-A1- 2004 077 773
US-B2- 9 890 276

- **EVONIK: "POLYVEST MA 75 Maleinsäureanhydrid-funktionalisiertes Flüssigpolybutadien", August 2011 (2011-08-01), pages 1 - 2, XP093120312, Retrieved from the Internet <URL:https://products.evonik.com/assets/88/69/TDS_POLYVEST_MA_75_DE_DE_Asset_1218869.pdf> [retrieved on 20240116]**

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Description

Technical Field

[0001] This application claims the benefit of priority based on Korean Patent Application No. 10-2020-0126958 filed on September 29, 2020.

[0002] The present invention relates to a resin composition having improved storage safety.

Background Art

[0003] A TIM (thermal interface material) material is usually prepared by blending a filler with a resin component.

[0004] Such a TIM material may be used in various applications, including battery modules and battery packs, and the like applied to electric vehicles.

[0005] As a resin component forming the TIM material, an epoxy resin component, a polyurethane resin component or a polysilicon component, and the like is generally used.

[0006] However, the resin components as above have a problem of poor storage stability in common.

[0007] For example, in the case of the epoxy resin component, an amide compound is usually used as a curing agent, where the amide compound reacts with moisture to rapidly increase the viscosity of the material. In addition, when materials of aliphatic series are applied, there is also a problem that impurities such as salts are generated on the surface during long-term storage.

[0008] In the case of a material of polyurethane series, an isocyanate compound is mainly used as a curing agent, and such a compound also easily reacts with moisture to cause a viscosity increase in the material.

[0009] In the case of a material of polysilicon series, it has poor miscibility with generally applied fillers, whereby the oil separation phenomenon, in which the filler and the resin component are separated, easily occurs, and it comprises a low molecular weight siloxane component, thereby easily causing contact failure issues and the like when applied to electrical products.

[0010] In the case of the filler included in the TIM material, it usually contains moisture itself in many cases, where the moisture contained in the filler in this way speeds up the above problems.

[0011] In addition, depending on the use of the TIM material, an excessive amount of filler is often blended in order to obtain high thermal conductivity, where the use of an excessive amount of filler makes it more difficult to solve the above problems.

[0012] WO 2020/100102 A2 discloses a curable composition comprising a polyol component, a functional butadiene component, and a thermally conductive filler.

Disclosure

Technical Problem

[0013] The present invention provides a resin composition. The present invention is intended to provide a resin composition that maintains a uniform mixing state without any oil separation phenomenon and the like even in a composition in which a filler is blended, and there is no viscosity increase even when stored for a long period of time.

[0014] The present invention is intended to provide a resin composition capable of achieving the above object even when it contains an excessive amount of a filler in a state that particularly contains moisture and has not undergone a separate surface treatment.

Technical Solution

[0015] Among the physical properties mentioned in this specification, when the measured temperature affects the result, the relevant physical property is a physical property measured at room temperature, unless otherwise specified. The term room temperature is a natural temperature without warming or cooling, which is usually one temperature within the range of about 10°C to 30°C, or a temperature of about 23°C or about 25°C or so. In addition, unless otherwise specified in this specification, the unit of temperature is °C.

[0016] Among the physical properties mentioned in this specification, when the measured pressure affects the result, the relevant physical property is a physical property measured at normal pressure, unless otherwise specified. The term normal pressure is a natural pressure without pressurization and depressurization, where about 1 atmosphere or so is usually referred to as the normal pressure.

[0017] The present invention relates to a resin composition used as a TIM (thermal interface material) material. The TIM (thermal interface material) material can be prepared by blending a filler with a resin component, and an epoxy resin

component, a polyurethane resin component or a polysilicon component, and the like can be generally used as the resin component forming such a TIM material. However, the resin components as above have poor storage stability in common. In particular, when the filler is included in the TIM material, it usually contains moisture itself in many cases, where the moisture contained in the filler in this way may further deteriorate the storage stability or keeping safety, and the like of the resin composition.

[0018] An aspect of the present invention is a two-component resin composition as defined in claim 1.

[0019] The two-component resin composition according to the present invention comprises a curing agent part (resin composition) which comprises a polymer compound having an organic acid anhydride functional group and a filler. When the curing agent part comprises a polymer compound having an organic acid anhydride functional group, it is possible to effectively prevent the curing agent part from reacting with moisture to increase the viscosity. The moisture reacting with the curing agent part may mean moisture existing outside the curing agent part , moisture included in the curing agent part, or moisture included in the filler.

[0020] As one example, the curing agent part may satisfy the following general formula 1.

[0021]

[General Formula 1]

$$V = V_2 / V_1 < 1.2$$

[0022] In General Formula 1, V is a viscosity change rate of a curing agent part, V_1 is an initial viscosity measured at room temperature and a shear rate of 2.4/s using Wells/Brookfield Cone & Plate's viscometer within 3 minutes from the time of preparing a curing agent part comprising a polymer compound having an organic acid anhydride functional group and a filler, and V_2 is a viscosity measured at room temperature and a shear rate of 2.4/s using Wells/Brookfield Cone & Plate's viscometer at the time of curing for 30 days from the time of preparing the curing agent part.

[0023] In General Formula 1 above, the curing agent part may be prepared by blending a filler with a polymer compound having an organic acid anhydride functional group and mixing them with a mixer.

[0024] As another example with regard to the measurement of V_1 in General Formula 1 above, it may be a viscosity measured within about 2.5 minutes or within about 2 minutes from the time of preparing the curing agent part.

[0025] As another example, the viscosity change rate (V) of the curing agent part in General Formula 1 above may be about 1.19 or less, 1.18 or less, 1.17 or less, 1.16 or less, or about 1.15 or less. The lower limit is not particularly limited, but may be about 1.00 or more, or more than about 1.00.

[0026] As one example, the organic acid anhydride functional group may be a functional group derived from benzoic anhydride, a functional group derived from phthalic anhydride or a functional group derived from maleic anhydride. The curing agent part comprising the polymer compound having an organic acid anhydride functional group of the above type has a low reaction with moisture, so that it may effectively prevent the viscosity increase of the curing agent part, and it may be advantageous to satisfy the aforementioned general formula 1.

[0027] As one example, the polymer compound is not particularly limited as long as it satisfies physical properties (molecular weight, glass transition temperature) to be described below, which may be, for example, a compound having an organic acid anhydride functional group substituted in a polybutadiene skeleton, a polyester skeleton or a polyether skeleton.

[0028] As one example, the polymer compound having the organic acid anhydride functional group may have a molecular weight (Mn) of 4,000 g/mol or less. As another example, the molecular weight (Mn) may be about 3,800 g/mol or less, 3,600 g/mol or less, 3,400 g/mol or less, 3,200 g/mol or less, or about 3,000 g/mol or less, and may be about 500 g/mol or more, 600 g/mol or more, 700 g/mol or more, 800 g/mol or more, 900 g/mol or more, or about 1,000 g/mol or more.

[0029] A curing agent part comprising a polymer compound having a molecular weight (Mn) exceeding 4,000 g/mol may have poor storage stability due to a high viscosity change rate, and may cause overload of the injection equipment when mixed with a main part to be described below and applied to a battery module or battery pack. Meanwhile, a curing agent part comprising a polymer compound having a molecular weight (Mn) of less than 500 g/mol has a very low viscosity, so that it takes a long time for the curing agent part to cure, and thus the productivity of the battery module or battery pack to which the curing agent part is applied may be lowered.

[0030] A curing agent part comprising a polymer compound having a molecular weight (Mn) in the above range may be more advantageous in satisfying the above-mentioned general formula 1 because it has low reactivity with moisture, may prevent overload of the injection equipment when mixed with the main part and applied to a battery module or battery pack, and may also improve the productivity of battery modules or battery packs.

[0031] In this specification, the "molecular weight" may be a number average molecular weight (Mn) measured using GPC (gel permeation chromatograph).

[0032] With regard to the measurement of the number average molecular weight, it can be measured using GPC under

the following conditions, where Agilent system's standard polystyrene can be used to prepare the calibration curve, thereby converting the measurement result.

<Molecular weight measurement conditions>

[0033]

Meter: Gel Permeation Chromatography (Waters Alliance System)

Column: PL Mixed B type

Detector: Refractive index detector

Column flow rate and solvent: 1 mL/min, Solvent: THF (tetrahydrofuran)

Analytical temperature and measurement volume: 40°C, 200 μL

[0034] As one example, the polymer compound having the organic acid anhydride functional group may have a glass transition temperature of 0°C or less. In another example, it may be about -5°C or less, about -10°C or less, about -15°C or less, about -20°C or less, about -25°C or less, about -30°C or less, about -35°C or less, about -40°C or less, about - 45°C or less, or about -50°C or less, and may be about -150°C or more, -140°C or more, - 130°C or more, -120°C or more, or about -100°C or more. The glass transition temperature may be measured using a DMA (dynamic mechanical analyzer) or differential scanning calorimetry (DSC), and the like.

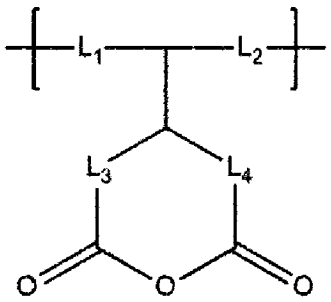
[0035] A curing agent part comprising a polymer compound having a glass transition temperature satisfying the above range may be more advantageous in satisfying the above-mentioned general formula 1 because it has low reactivity with moisture.

[0036] As one example, the polymer compound may have an acid value according to DIN EN ISO 2114 in the range of 50 mgKOH/g to 120 mgKOH/g. In another example, the acid value may be about 52 mgKOH/g or more, 54 mgKOH/g or more, 56 mgKOH/g or more, 58 mgKOH/g or more, or about 60 mgKOH/g or more, and may be about 118 mgKOH/g or less, 116 mgKOH/g or less, 114 mgKOH/g or less, 112 mgKOH/g or less, or about 110 mgKOH/g or less.

[0037] The curing agent part including the polymer compound satisfying the acid value in the above range has low reactivity with water, so that it may effectively prevent the viscosity increase of the curing agent part, and it may be more advantageous to satisfy the above-mentioned general formula 1.

[0038] As one example, the polymer compound may comprise a polymerization unit of Formula 1 below.

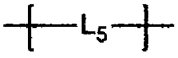
[Formula 1]



[0039] In Formula 1, L_1 is a single bond or an alkenylene group with 2 to 4 carbon atoms, L_2 is a single bond or an alkylene group with 1 to 4 carbon atoms, and L_3 and L_4 are each independently a single bond or an alkylene group with 1 to 4 carbon atoms.

[0040] In addition, the polymer compound may further comprise a polymerization unit of Formula 2 below.

[Formula 2]



[0041] In Formula 2, L_5 is an alkenylene group with 2 to 4 carbon atoms or an alkylene group with 1 to 4 carbon atoms substituted with an alkenyl group with 2 to 4 carbon atoms.

[0042] The polymer compound comprising the polymerization unit of Formula 1 and the polymerization unit of Formula 2 has low reactivity with water, so that it may more effectively prevent the viscosity increase of the resin composition, and may be more advantageous in satisfying the aforementioned general formula 1.

[0043] The curing agent part according to the present invention comprises a filler. The filler may be a thermally conductive filler. In the present invention, the term thermally conductive filler may mean a filler made of a material having a thermal conductivity of about 3 W/mK or more, 5 W/mK or more, 10 W/mK or more, or about 15 W/mK or more. Specifically, the thermal conductivity of the thermally conductive filler may be about 400 W/mK or less, 350 W/mK or less, or about 300 W/mK or less. The type of the usable thermally conductive filler is not particularly limited, but may be an inorganic filler when considering insulation and the like together. For example, ceramic particles such as aluminum oxide (alumina: Al_2O_3), aluminum nitride (AlN), boron nitride (BN), silicon nitride (Si_3N_4), silicon carbide (SiC), beryllium oxide (BeO), zinc oxide (ZnO), magnesium oxide (MgO) or boehmite may be used. The shape or ratio of the filler is not particularly limited, which may be appropriately adjusted in consideration of the viscosity of the curing agent part, or the viscosity change rate, dispersibility or storage stability, and the like of the curing agent part. In general, as the size of the filler increases, the viscosity of the composition comprising the same increases, and the sedimentation possibility of the filler increases. Also, the smaller the size, the higher the thermal resistance tends to be. Therefore, in consideration of such points, a filler of an appropriate type and size may be selected, and if necessary, two or more fillers may also be used together. In addition, it is advantageous to use a spherical filler in consideration of the amount to be filled, but a filler having a needle shape or a plate shape may also be used in consideration of network formation or conductivity, and the like.

[0044] In one example, the filler may have an average particle diameter in a range of about 0.001 μm to 80 μm. In another example, the average particle diameter of the filler may be about 0.01 μm or more, 0.1 μm or more, 0.5 μm or more, 1 μm or more, 2 μm or more, 3 μm or more, 4 μm or more, 5 μm or more, or about 6 μm or more. In another example, the average particle diameter of the filler may be about 75 μm or less, 70 μm or less, 65 μm or less, 60 μm or less, 55 μm or less, 50 μm or less, 45 μm or less, 40 μm or less, 35 μm or less, 30 μm or less, 25 μm or less, 20 μm or less, 15 μm or less, 10 μm or less, or about 5 μm or less.

[0045] In order to obtain excellent heat dissipation performance, it may be considered that a high content of the thermally conductive filler is used. The filler is included in the curing agent part in an amount of 75 weight% to 92 weight%, relative to 100 weight% of the curing agent part. Specifically, the content of the filler may be 76 weight% or more, 77 weight% or more, 78 weight% or more, 79 weight% or more, or 80 weight% or more, and 91.5 weight% or less, 91 weight% or less, 90.5 weight% or less, or 90 weight% or less, relative to 100 weight% of the curing agent part. The desired physical properties such as thermal conductivity and storage stability may be secured within the ratio range of the filler.

[0046] The moisture content of the filler in the curing agent part is 10 ppm to 3,000 ppm, and may be about 100 ppm or more, or about 1,000 ppm or more, and 2,900 ppm or less, or about 2,800 ppm or less. The moisture content of the filler can be measured with a Karl Fischer titrator (KR831) under the conditions of 10% relative humidity and a drift of 5.0 or less. At this time, the moisture content may be an average moisture content with respect to all fillers used in the curing agent part.

[0047] The curing agent part lowers the moisture content of the filler in order to lower the viscosity change rate by moisture, or has required a pretreatment process of the filler, such as surface treatment of the filler, in order to prevent oil separation from occurring. The curing agent part comprising the polymer compound having an organic acid anhydride functional group according to the present invention has low reactivity with moisture even when the moisture content contained in the filler is within the range of 10 ppm to 3,000 ppm, so that the viscosity change of the curing agent part is not large, and thus the storage stability of the curing agent part can be greatly improved. In addition, since the pretreatment process of the filler is not necessarily accompanied, the manufacturing cost can be reduced and it has an effect of improving the manufacturing processability.

[0048] In addition to the above, various types of fillers may be used. For example, in order to secure insulating properties of a cured product in which the curing agent part is cured, the use of a carbon filler such as graphite may be considered. Alternatively, for example, a filler such as fumed silica, clay, calcium carbonate ($CaCO_3$), zinc oxide (ZnO) or aluminum hydroxide ($Al(OH)_3$) may be used. The type or content ratio of such a filler is not particularly limited, which may be selected in consideration of viscosity, viscosity change rate, sedimentation possibility, thixotropy, insulation, filling effects or storage stability, and the like of the curing agent part .

[0049] The curing agent part does not comprise an amine catalyst or an isocyanate compound. Excluded amine catalysts are exemplified by 2,4,6-tris (dimethylaminomethyl)phenol, N,N-dimethylpropionamide, imidazole, 1,4-diaza-bicyclo(2,2,2)octane, bis(2-dimethylaminoethyl)ether, trimethylaminoethylethanolamine, pentamethyldiethylenetriamine, N,N'-dimethylethanolamine, dimethylaminopropylamine, N-ethylmorpholine, N,N-dimethylaminoethylmorpholine, N,N-dimethylcyclohexylamine and 2-methyl-2-azanorbornene. Excluded isocyanate compounds are exemplified by known aromatic isocyanate compounds and non-aromatic isocyanate compounds.

[0050] The polymer compound having an organic acid anhydride functional group may react with moisture in the presence of an amine catalyst, which may cause a viscosity increase of the curing agent part. Therefore, the storage

stability of the curing agent part may be deteriorated. In addition, when the isocyanate compound is present in the curing agent part, it may react with moisture to cause a viscosity increase of the curing agent part as well.

[0051] When the curing agent part does not comprise any amine catalyst or any isocyanate compound, the moisture reactivity of the curing agent part is significantly lowered, so that the storage stability of the curing agent part may be improved.

[0052] The two-component resin composition according to the present invention comprises a main part including a main resin and a filler, and a curing agent part. The curing agent part is as described above, that is, the curing agent part including a polymer compound having an organic acid anhydride functional group and a filler.

[0053] As one example, the filler included in the main part may use the above-described thermally conductive filler.

[0054] As the main resin, an ester polyol resin is used. The ester polyol may have amorphousness or a polyol having sufficiently low crystallinity.

[0055] In this specification, the "amorphousness" means a case in which the crystallization temperature (Tc) and the melting temperature (Tm) are not observed in a DSC (differential scanning calorimetry) analysis to be described below. At this time, the DSC analysis may be performed within the range of -80 to 60°C at a rate of 10°C/min, which may be made, for example, in a manner that the temperature is raised from 25°C to 60°C at the above rate, and then reduced to -80°C again and raised to 60°C again. In addition, here, the "sufficiently low crystallinity" means a case in which the melting point (Tm) observed in the DSC analysis is less than 15°C, about 10°C or less, 5°C or less, 0°C or less, -5°C or less, -10°C or less, or about -20°C or less or so. At this time, the lower limit of the melting point is not particularly limited, but the melting point may be, for example, about -80°C or more, -75°C or more, or about -70°C or more. When the polyol has crystallinity or strong (room temperature) crystallinity such as the case that the melting point range is not satisfied, the viscosity difference according to the temperature is likely to increase, so that the manufacturing processability may be deteriorated.

[0056] In one example, as the ester polyol, for example, a carboxylic acid polyol or a caprolactone polyol may be used.

[0057] The carboxylic acid polyol may be formed by reacting components comprising a carboxylic acid and a polyol (e.g., a diol or a triol, etc.), and the caprolactone polyol may be formed by reacting components comprising caprolactone and a polyol (e.g., a diol or a triol). At this time, the carboxylic acid may be a dicarboxylic acid.

[0058] The main part and the curing agent part may react at room temperature to be cured. Specifically, the ester polyol resin in the main part and the polymer compound having an organic acid anhydride functional group in the curing agent part may react at room temperature to be cured.

[0059] The curing reaction may be assisted by, for example, a catalyst. Accordingly, the two-component resin composition may include all states that the main resin (polyol) and the curing agent (polymer compound having an organic acid anhydride functional group) are separated, mixed, or reacted.

[0060] The main part comprises an amine catalyst to accelerate the reaction between the main resin and the curing agent. For example, at least one of 2,4,6-tris(dimethylaminomethyl)phenol, N,N-dimethylpropionamide, imidazole, 1,4-diazabicyclo(2,2,2)octane, bis(2-dimethylaminoethyl)ether, trimethylaminoethylethanolamine, pentamethyldiethylene-triamine, N,N'-dimethylethanolamine, dimethylaminopropylamine, N-ethylmorpholine, N,N-dimethylaminoethylmorpholine, N,N-dimethylcyclohexylamine and 2-methyl-2-azanorbornene may be used.

[0061] The catalyst may be included in the main part in a range of about 0.01 weight% to 5 weight% relative to 100 weight% of the two-component resin composition. When the catalyst is included in the main part within the above range, it is advantageous to improve the storage stability of the two-component resin composition. In addition, when the two-component resin composition is injected, the two-component resin composition has a low viscosity, so that the manufacturing processability can be improved, and after injection, the curing rate of the two-component resin composition is accelerated, so that the process tact time of the battery module can be improved.

[0062] The two-component resin composition further comprises a dispersant in at least one of the main part and the curing agent part. As the type of dispersant, a copolymer phosphoric acid-based dispersant containing a propylene glycol methyl ether group is used in consideration of the desired viscosity and storage stability of the resin composition. As one example, when the dispersant is comprised in both the main part and the curing agent part, regarding the content of the dispersant, the dispersant included in the main part may be included in the range of 0.05 weight% to 10 weight% relative to 100 weight% of the two-component resin composition, and the dispersant included in the curing agent part may be included in the range of 0.01 weight% to 5 weight% relative to 100 weight% of the two-component resin composition.

[0063] When such a phosphoric acid-based dispersant is used in the above-described content ratio in the main part and the curing agent part, the viscosity change of the two-component resin composition can be lowered and the oil separation can be more effectively prevented.

[0064] As one example, the two-component resin composition may form a cured product having a thermal conductivity of 2.0 W/mK or more as measured according to ISO22007-2 standard. As another example, it may be about 2.5 W/mK or more, 3.0 W/mK or more, 3.5 W/mK or more, or 4.0 W/mK or more, and may be about 50 W/mK or less, 45 W/mK or less, 40 W/mK or less, 35 W/mK or less, 30 W/mK or less, 25 W/mK or less, 20 W/mK or less, 15 W/mK or less, 10 W/mK or less, 5 W/mK or less, 4.5 W/mK or less, or about 4.0 W/mK. When the cured product of the two-component resin composition satisfies the thermal conductivity within the above range, the heat dissipation performance of the battery module or battery

pack to which the two-component resin composition is applied may be improved.

[0065] The present application may also relate to a battery module. The battery module comprises a module case and a plurality of battery cells accommodated in an inner space of the module case. The number of battery cells accommodated in the module case is not particularly limited as it is adjusted according to the use or the like. The battery cells accommodated in the module case may be electrically connected to each other.

[0066] The battery module according to the present invention also comprises a resin layer in contact with the plurality of battery cells and the module case. The resin layer may be a cured layer of the above-described two-component resin composition.

[0067] Two or more battery modules as described above may be included in a battery pack. In the battery pack, the battery modules may be electrically connected to each other. A method of configuring a battery pack by electrically connecting two or more battery modules is not particularly limited, and any known method may be applied.

Advantageous Effects

[0068] The curing agent part according to the present invention can maintain a uniform mixing state without any oil separation phenomenon even though a filler is blended, and can prevent or reduce viscosity increase even during long-term storage. In addition, the curing agent part according to the present invention may achieve the above effects even when it comprises an excessive amount of a filler in a state in which it contains moisture and is not subjected to a separate surface treatment.

Mode for Invention

[0069] Hereinafter, the present invention will be described in detail through Examples.

Storage stability evaluation

[0070] Storage stability evaluation was performed using the curing agent part (resin composition) prepared in Examples and Comparative Examples. When the value of the following general formula 1 was 1.2 or more, it was assumed that there was no storage stability.

[General Formula 1]

$$V = V_2 / V_1 < 1.2$$

[0071] In General Formula 1, V is a viscosity change rate of a resin composition, V_1 is an initial viscosity measured at room temperature and a shear rate of 2.4/s using Wells/Brookfield Cone & Plate's viscometer within 3 minutes from the time of preparing a resin composition comprising a polymer compound having an organic acid anhydride functional group and a filler, and V_2 is a viscosity measured at room temperature and a shear rate of 2.4/s using Wells/Brookfield Cone & Plate's viscometer at the time of curing for 30 days from the time of preparing the resin composition.

[0072] In addition, it was determined that there was no storage stability when white crystals were formed due to salt generation or oil separation was observed within 3 days from the time the resin composition was prepared.

[Evaluation criteria]

[0073]

O: The value of General Formula 1 above is less than 1.2, no salt generation, no oil separation phenomenon is observed within 3 days (if all are applicable)

X: The value of General Formula 1 above is 1.2 or more, salt generation presence, oil separation phenomenon is observed within 3 days (if any of these is applicable)

Thermal conductivity

[0074] Thermal conductivity was measured by a hot disk method according to ISO22007-2 standard using a cured product of a two-component resin composition prepared from the main part and the curing agent part prepared in Examples and Comparative Examples by means of a static mixer. At this time, the two-component resin composition was

prepared so that the volume ratio of the main part and the curing agent part was 1:1 or so.

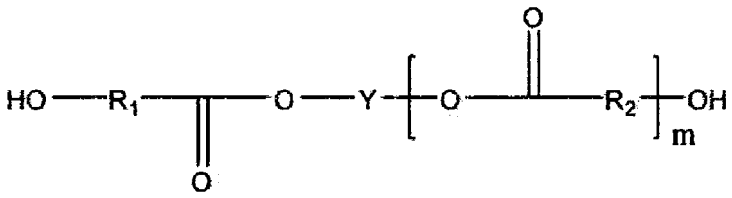
[0075] Specifically, regarding the thermal conductivity measurement, the thermal conductivity can be measured in the through plane direction by placing the cured product of the two-component resin composition in a mold having a thickness of about 5 mm or so and using a hot disk device. As stipulated in the above standard (ISO 22007-2), the hot disk device is a device that can identify the thermal conductivity by measuring the temperature change (electrical resistance change) as a sensor with a nickel wire of a double spiral structure is heated, and such a thermal conductivity was measured according to the standard.

Example

Main part:

[0076] As the main resin, a caprolactone polyol represented by the following formula 3 was used.

[Formula 3]



[0077] In Formula 3, m is a number within a range of 1 to 3, R_1 and R_2 are each alkylene with 4 carbon atoms, and Y is a 1,4-butanediol unit.

[0078] As a filler, alumina was used, and one without any treatment on the particle surface was used as it was.

[0079] Daejungchem's DMP-30 (2,4,6-tris(dimethylaminomethyl)phenol) was used as a catalyst, and BYK's BKY-111 was used as a dispersant.

[0080] The main part was prepared by mixing the caprolactone polyol, the filler, the catalyst and the dispersant in a weight ratio of 10.36:89:0.34:0.3 (polyol: filler: catalyst: dispersant).

Curing agent part (resin composition):

[0081] As a polymer compound having an organic acid anhydride functional group, EVONIK's POLYVEST MA 75 was used (number average molecular weight: 3,000 g/mol, glass transition temperature: -95°C, acid value: 70-90 mgKOH/g).

[0082] As a filler, alumina was used, and one without any treatment on the particle surface was used as it was.

[0083] As a dispersant, BYK's BKY-118 was used.

[0084] The curing agent part was prepared by mixing the polymer compound having the organic acid anhydride functional group, the filler and the dispersant in a weight ratio of 10.85:89:0.15 (polymer compound: filler: dispersant).

[0085] The mixing during the preparation of the main part and the curing agent part was performed with a planetary mixer.

Comparative Example 1

[0086] Main part: It was prepared by mixing the same components in the same ratios as in the Example, except that a tin catalyst (Sigma-Aldrich's DBTDL) was used instead of the amine catalyst.

[0087] Curing agent part: It was prepared by mixing the same components in the same ratios as in the Example, except that a polyisocyanate (HDI, hexamethylene diisocyanate) was used instead of the polymer compound of the Example, and as the dispersant, BYK's BKY-111 was used.

Comparative Example 2

[0088] Main part: It was prepared by mixing the same components in the same ratios as in the Example, except that an epoxy resin (Kukdo Chemical's YH-300) was used instead of the polyol resin as the main resin and as the dispersant, BYK's BKY-102 was used.

[0089] Curing agent part: It was prepared by mixing the same components in the same ratios as in the Example, except that an amide (Kukdo Chemical's G-A0432) was used instead of the polymer compound of the Example, and as the

dispersant, BYK's BKY-102 was used.

Comparative Example 3

[0090] Main part: It was prepared by mixing the same components in the same ratios as in Comparative Example 2.

[0091] Curing agent part: It was prepared by mixing the same components in the same ratios as in the Example, except that an aliphatic amine (Kukdo Chemical' KH-8108) was used instead of the polymer compound of the Example, and as the dispersant, BYK's BKY-102 was used.

Comparative Example 4

[0092] Main part: It was prepared by mixing the same components in the same ratios as in the Example, except that as the main resin, siloxane (KCC's SF3000E, SF6003P, Modifier715 and Inhibitor600) was used, and as the dispersant, BYK's BYK-1799 was used.

[0093] Curing agent part: It was prepared by mixing the same components in the same ratios as in the Example, except that siloxane (KCC's SF3000E) was used instead of the polymer compound of the Example, and as the dispersant, BYK's BYK-1799 was used.

[0094] The storage stability evaluation and thermal conductivities measured for the Example and Comparative Examples were summarized and described in Table 1 below.

[Table 1]

	Storage stability	Thermal conductivity (W/mK)
Example	O	2.6
Comparative Example 1	X (viscosity increase more than twice for 1 month)	2.8
Comparative Example 2	X (viscosity increase 1.5 times or more for 1 month)	3.0
Comparative Example 3	X (white crystal generation due to salt generation)	3.0
Comparative Example 4	X (oil separation after 3 days)	2.6

[0095] From the results in Table 1, in the case of the Example comprising the polymer compound having an organic acid anhydride functional group in the resin composition, it can be seen that it has excellent storage stability even without performing the moisture content control of the filler or the surface treatment of the filler. In addition, the cured product of the two-component resin composition comprising the resin composition in the curing agent part had an excellent thermal conductivity of 2.0 W/mK or more.

[0096] In comparison, in the case of Comparative Example 1 comprising the isocyanate instead of the polymer compound having an organic acid anhydride functional group in the resin composition or in the case of Comparative Example 2 comprising the amide instead of the polymer compound having an organic acid anhydride functional group in the resin composition, it can be seen that the viscosity change rate exceeds 1.2, whereby the storage stability is deteriorated. Also, in the case of Comparative Example 3 comprising the aliphatic amine instead of the polymer compound having an organic acid anhydride functional group in the resin composition, white crystals were produced due to salt generation. In addition, in the case of Comparative Example 4 comprising the siloxane instead of the polymer compound having an organic acid anhydride functional group in the resin composition, it can be seen that the oil separation phenomenon is observed within 3 days from the time the resin composition is prepared, whereby the storage stability is deteriorated.

Claims

1. A two-component resin composition comprising

(i) a main part comprising a main resin and a filler; and
(ii) a curing agent part comprising a polymer compound having an organic acid anhydride functional group and a filler;
wherein

the main resin is an ester polyol resin;

the filler is contained in the curing agent part in an amount of 75 to 92 weight% based on 100 weight% of the curing agent part;
the filler in the curing agent part has a moisture content in the range of 10 ppm to 3,000 ppm, as determined using the method set out in the description;
at least one of the main part and the curing agent part further comprises a copolymer phosphoric acid-based dispersant containing a propylene glycol methyl ether group;
the main part further comprises an amine catalyst; and
the curing agent part does not comprise an amine catalyst or an isocyanate compound.

2. The two-component resin composition according to claim 1, wherein the organic acid anhydride functional group is a functional group derived from benzoic anhydride, a functional group derived from phthalic anhydride, or a functional group derived from maleic anhydride.

3. The two-component resin composition according to claim 1, wherein the polymer compound has an organic acid anhydride functional group substituted in a polybutadiene skeleton, a polyester skeleton or a polyether skeleton.

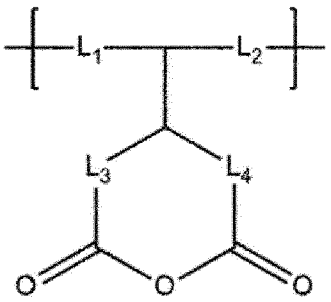
4. The two-component resin composition according to claim 1, wherein the polymer compound has a number average molecular weight (Mn) of 4,000 g/mol or less, as determined using the method set out in the description.

5. The two-component resin composition according to claim 1, wherein the polymer compound has a glass transition temperature (Tg) of 0°C or less, as determined using the method set out in the description.

6. The two-component resin composition according to claim 1, wherein the polymer compound has an acid value according to DIN EN ISO 2114 in a range of 50 mgKOH/g to 120 mgKOH/g.

7. The two-component resin composition according to claim 1, wherein the polymer compound comprises a polymerization unit represented by the following formula 1:

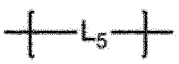
[Formula 1]



wherein L_1 is a single bond or an alkenylene group with 2 to 4 carbon atoms, L_2 is a single bond or an alkylene group with 1 to 4 carbon atoms, and L_3 and L_4 are each independently a single bond or an alkylene group with 1 to 4 carbon atoms.

8. The two-component resin composition according to claim 1, wherein the polymer compound further comprises a polymerization unit represented by the following formula 2:

[Formula 2]



wherein L_5 is an alkenylene group with 2 to 4 carbon atoms or an alkylene group with 1 to 4 carbon atoms substituted with an alkenyl group with 2 to 4 carbon atoms.

9. The two-component resin composition according to claim 1, wherein the filler in the curing agent part is fumed silica, clay, calcium carbonate ($CaCO_3$), aluminum oxide (Al_2O_3), aluminum nitride (AlN), boron nitride (BN), silicon nitride

(Si_3N_4), silicon carbide (SiC), beryllium oxide (BeO), zinc oxide (ZnO), aluminum hydroxide ($Al(OH)_3$), boehmite or a carbon filler.

10. The two-component resin composition according to claim 1, wherein the ester polyol is a carboxylic acid polyol.

11. The two-component resin composition according to claim 1, wherein the ester polyol is a caprolactone polyol.

12. The two-component resin composition according to claim 10 or 11, wherein the ester polyol has amorphousness, meaning that a crystallization temperature (Tc) and a melting temperature (Tm) are not observed in differential scanning calorimetry performed within the temperature range of -80°C to 60°C at a rate of 10°C/min.

13. The two-component resin composition according to claim 10 or 11, wherein the ester polyol has a melting point of less than 15°C, as determined using the method set out in the description.

14. A battery module comprising

a module case;
a plurality of battery cells accommodated in an inner space of the module case; and
a resin layer in contact with the plurality of battery cells and the module case;
wherein the resin layer is a cured layer of the two-component resin composition of any of claims 1-13.

Patentansprüche

1. Zweikomponentige Harzzusammensetzung, umfassend

(i) einen Hauptteil, der ein Hauptharz und einen Füllstoff umfasst, und
(ii) ein Härtungsmittelteil, der eine Polymerverbindung mit einer funktionellen organischen Säureanhydridgruppe und einen Füllstoff umfasst,
wobei

das Hauptharz ein Esterpolyolharz ist,
der Füllstoff in dem Härtungsmittelteil in einer Menge von 75 bis 92 Gew.-%, bezogen auf 100 Gew.-% des Härtungsmittelteils, enthalten ist,
der Füllstoff in dem Härtungsmittelteil einen Feuchtigkeitsgehalt im Bereich von 10 ppm bis 2.000 ppm, bestimmt unter Verwendung des in der Beschreibung dargelegten Verfahrens, aufweist,
mindestens einer von dem Hautteil und dem Härtungsmittelteil ferner ein Copolymer-Phosphorsäurebasiertes Dispergiermittel umfasst, das eine Propylenglykolmethylethergruppe enthält,
der Hauptteil ferner einen Aminkatalysator umfasst und
der Härtungsmittelteil weder einen Aminkatalysator noch eine Isocyanatverbindung umfasst.

2. Zweikomponentige Zusammensetzung gemäß Anspruch 1, wobei die funktionelle organische Säureanhydridgruppe eine von Benzoesäureanhydrid abgeleitete funktionelle Gruppe, eine von Phthalsäureanhydrid abgeleitete funktionelle Gruppe oder eine von Maleinsäureanhaydrid abgeleitete funktionelle Gruppe ist.

3. Zweikomponentige Zusammensetzung gemäß Anspruch 1, wobei die Polymerverbindung eine funktionelle organische Säureanhydridgruppe, substituiert in ein Polybutadiengerüst, ein Polyestergerüst oder ein Polyethergerüst, aufweist.

4. Zweikomponentige Zusammensetzung gemäß Anspruch 1, wobei die Polymerverbindung ein zahlenmittleres Molekulargewicht (Mn) von 4.000 g/mol oder weniger, bestimmt unter Verwendung des in der Beschreibung dargelegten Verfahrens, aufweist.

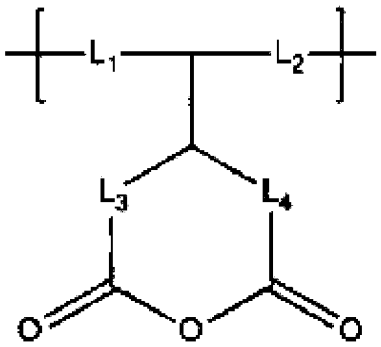
5. Zweikomponentige Zusammensetzung gemäß Anspruch 1, wobei die Polymerverbindung eine Glasübergangstemperatur (Tg) von 0°C oder weniger, bestimmt unter Verwendung des in der Beschreibung dargelegten Verfahrens, aufweist.

6. Zweikomponentige Zusammensetzung gemäß Anspruch 1, wobei die Polymerverbindung einen Säurewert gemäß

DIN EN ISO 2114 in einem Bereich von 50 mgKOH/g bis 120 mgKOH/g aufweist.

7. Zweikomponentige Zusammensetzung gemäß Anspruch 1, wobei die Polymerverbindung eine durch die folgende Formel 1 dargestellte Polymerisationseinheit umfasst:

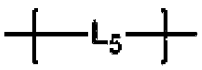
[Formel 1]



worin L_1 eine Einfachbindung oder eine Alkenylengruppe mit 2 bis 4 Kohlenstoffatomen ist, L_2 eine Einfachbindung oder eine Alkylengruppe mit 1 bis 4 Kohlenstoffatomen ist und L_3 und L_4 jeweils unabhängig eine Einfachbindung oder eine Alkylengruppe mit 1 bis 4 Kohlenstoffatomen sind.

8. Zweikomponentige Harzzusammensetzung gemäß Anspruch 1, wobei die Polymerverbindung ferner eine durch die folgende Formel 2 dargestellte Polymerisationseinheit umfasst:

[Formel 2]



worin L_5 eine Alkenylengruppe mit 2 bis 4 Kohlenstoffatomen oder eine Alkylengruppe mit 1 bis 4 Kohlenstoffatomen, substituiert mit einer Alkenylgruppe mit 2 bis 4 Kohlenstoffatomen, ist.

9. Zweikomponentige Harzzusammensetzung gemäß Anspruch 1, wobei der Füllstoff in dem Härtungsmittelteil pyrogenes Siliciumdioxid, Ton, Calciumcarbonat ($CaCO_3$), Aluminiumoxid (Al_2O_3), Aluminiumnitrid (AlN), Bornitrid (BN), Siliciumnitrid (Si_3N_4), Silciumcarbid (SiC), Berylliumoxid (BeO), Zinkoxid (ZnO), Aluminiumhydroxid ($Al(OH)_3$), Böhmit oder ein Kohlenstofffüllstoff ist.

10. Zweikomponentige Harzzusammensetzung gemäß Anspruch 1, wobei das Esterpolyol ein Carbonsäurepolyol ist.

11. Zweikomponentige Harzzusammensetzung gemäß Anspruch 1, wobei das Esterpolyol ein Caprolactonpolyol ist.

12. Zweikomponentige Harzzusammensetzung gemäß Anspruch 10 oder 11, wobei das Esterpolyol Amorphität aufweist, was bedeutet, dass eine Kristallisationstemperatur (Tc) und eine Schmelztemperatur (Tm) in einer Differential-Scanning-Kalorimetrie, durchgeführt in einem Temperaturbereich von -80°C bis 60°C bei einer Geschwindigkeit von 10°C/min, nicht beobachtet werden.

13. Zweikomponentige Harzzusammensetzung gemäß Anspruch 10 oder 11, wobei das Esterpolyol einen Schmelzpunkt von weniger als 15°C, bestimmt unter Verwendung des in der Beschreibung dargelegten Verfahrens, aufweist.

14. Batteriemodul, umfassend

ein Modulgehäuse,
mehrere Batteriezellen, die in einem Innenraum des Modulgehäuses untergebracht sind, und
eine Harzschicht, die in Kontakt mit den mehreren Batteriezellen und dem Modulgehäuse steht,

wobei die Harzschicht eine gehärtete Schicht aus der zweikomponentigen Harzzusammensetzung nach einem der Ansprüche 1-13 ist.

Revendications

1. Composition de résine à deux composants comprenant

(i) une partie principale comprenant une résine principale et une charge ; et
(ii) une partie d'agent de durcissement comprenant un composé polymère présentant un groupe fonctionnel anhydride d'acide organique et une charge ;
dans laquelle

la résine principale est une résine polyol ester ;
la charge est contenue dans la partie d'agent de durcissement en une quantité de 75 à 92 % en poids sur la base de 100 % en poids de la partie d'agent de durcissement ;
la charge dans la partie d'agent de durcissement présente une teneur en humidité dans la plage de 10 ppm à 3 000 ppm, comme déterminé en utilisant le procédé exposé dans la description ;
au moins une partie parmi la partie principale et la partie d'agent de durcissement comprend en outre un dispersant à base d'acide phosphorique de type copolymère contenant un groupe éther méthylique de propylèneglycol ;
la partie principale comprend en outre un catalyseur aminé ; et
la partie d'agent de durcissement ne comprend pas de catalyseur aminé ou de composé isocyanate.

2. Composition de résine à deux composants selon la revendication 1, dans laquelle le groupe fonctionnel anhydride d'acide organique est un groupe fonctionnel dérivé de l'anhydride benzoïque, un groupe fonctionnel dérivé de l'anhydride phtalique, ou un groupe fonctionnel dérivé de l'anhydride maléique.

3. Composition de résine à deux composants selon la revendication 1, dans laquelle le composé polymère présente un groupe fonctionnel anhydride d'acide organique substitué dans un squelette polybutadiène, un squelette polyester ou un squelette polyéther.

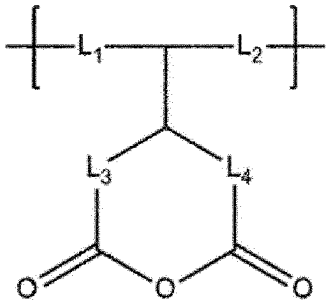
4. Composition de résine à deux composants selon la revendication 1, dans laquelle le composé polymère présente une masse moléculaire moyenne en nombre (Mn) de 4 000 g/mol ou inférieure, comme déterminé en utilisant le procédé exposé dans la description.

5. Composition de résine à deux composants selon la revendication 1, dans laquelle le composé polymère présente une température de transition vitreuse (Tg) de 0 °C ou inférieure, comme déterminé en utilisant le procédé exposé dans la description.

6. Composition de résine à deux composants selon la revendication 1, dans laquelle le composé polymère présente un indice d'acide selon la norme DIN EN ISO 2114 dans une plage de 50 mgKOH/g à 120 mgKOH/g.

7. Composition de résine à deux composants selon la revendication 1, dans laquelle le composé polymère comprend un motif de polymérisation représenté par la formule 1 suivante :

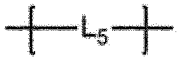
[Formule 1]



dans laquelle L_1 est une liaison simple ou un groupe alcénylène présentant 2 à 4 atomes de carbone, L_2 est une liaison simple ou un groupe alkylène présentant 1 à 4 atomes de carbone, et L_3 et L_4 sont chacun indépendamment une liaison simple ou un groupe alkylène présentant 1 à 4 atomes de carbone.

8. Composition de résine à deux composants selon la revendication 1, dans laquelle le composé polymère comprend en outre un motif de polymérisation représenté par la formule 2 suivante :

[Formule 2]



dans laquelle L_5 est un groupe alcénylène présentant 2 à 4 atomes de carbone ou un groupe alkylène présentant 1 à 4 atomes de carbone substitué par un groupe alcényle présentant 2 à 4 atomes de carbone.

9. Composition de résine à deux composants selon la revendication 1, dans laquelle la charge dans la partie d'agent de durcissement est de la silice pyrogénée, de l'argile, du carbonate de calcium ($CaCO_3$), de l'oxyde d'aluminium (Al_2O_3), du nitrure d'aluminium (AlN), du nitrure de bore (BN), du nitrure de silicium (Si_3N_4), du carbure de silicium (SiC), de l'oxyde de béryllium (BeO), de l'oxyde de zinc (ZnO), de l'hydroxyde d'aluminium ($Al(OH)_3$), de la boehmite ou une charge de carbone.

10. Composition de résine à deux composants selon la revendication 1, dans laquelle le polyol ester est un polyol d'acide carboxylique.

11. Composition de résine à deux composants selon la revendication 1, dans laquelle le polyol ester est un polyol de caprolactone.

12. Composition de résine à deux composants selon la revendication 10 ou la revendication 11, dans laquelle le polyol ester présente une amorphie, signifiant qu'une température de cristallisation (Tc) et une température de fusion (Tm) ne sont pas observées en calorimétrie différentielle à balayage réalisée dans la plage de température de -80 °C à 60 °C à une vitesse de 10 °C/min.

13. Composition de résine à deux composants selon la revendication 10 ou la revendication 11, dans laquelle le polyol ester présente un point de fusion inférieur à 15 °C, comme déterminé en utilisant le procédé exposé dans la description.

14. Module de batterie comprenant

un boîtier de module ;
une pluralité de cellules de batterie logées dans un espace interne du boîtier de module ; et
une couche de résine en contact avec la pluralité de cellules de batterie et le boîtier de module ;
dans lequel la couche de résine est une couche durcie de la composition de résine à deux composants selon l'une quelconque des revendications 1 à 13.

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- KR 1020200126958 **[0001]**
- WO 2020100102 A2 **[0012]**